MAR13-2012-000442
Abstract for an Invited Paper
for the MAR13 Meeting of
the American Physical Society

A Simple Model for Yielding and Strain Hardening in Glassy Polymers

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Strain hardening has long been an observed feature of polymer glasses in extension; explanations to date have often been phenomenological. Ediger and coworkers (Lee et al. *Science* 323, 231, 2009) have shown in experiments on PMMA glasses that, in addition to strain hardening, polymeric glasses show a remarkable non-monotonicity in the segmental relaxation time both in loading and unloading of stress. Here, we develop a simple constitutive equation that combines recent theories for yielding in simple glasses (Brader et al. PNAS, 106, 15186, 2009) to represent local segmental modes in the polymer, with a dumbbell model for the slow polymer relaxation modes. For a polymer glass under uniaxial loading, the model predicts that the liquefaction of the segmental modes permits strain hardening of the polymer modes to emerge, and once this emerges, it slows the deformation of the material under constant load enough to partially re-vitrify the segmental modes even though the sample remains under stress. In this way, the observed non-monotonicity in the segmental relaxation modes is produced. We show the extension of the work to simple shearing flows, and make (as yet) untested predictions about segmental relaxation rates in shear flows. We also show how to extend the model to include Rouse chain dynamics in place of the over-simplified dumbbell.